

Quantum phase transitions of the $S = 1$ Shastry-Sutherland model

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We investigate quantum phase transitions of the two-dimensional $S = 1$ Shastry-Sutherland model, which is characterized by the frustrated orthogonal-dimer structure, by means of the exact diagonalization method and the Ising-type series expansion method. We clarify how distinct spin gap phases realized in the chain system are adiabatically connected to those in the two-dimensional Shastry-Sutherland model. The effect of single-ion anisotropy is also discussed.

KEYWORDS: Shastry-Sutherland model, Valence Bond Solid, exact diagonalization, Ising expansion

1. Introduction

Frustrated spin systems have attracted much theoretical attention over many years. Interesting experimental realizations have been recently found in the transition-metal oxides $\text{SrCu}_2(\text{BO}_3)_2$ ^{1,2)} and $\text{Nd}_2\text{BaZnO}_5$,³⁾ where the magnetic ions Cu^{2+} and Nd^{3+} sit on the orthogonal-dimer structure (see Fig. 1). In the compound $\text{SrCu}_2(\text{BO}_3)_2$, a dimer singlet phase is realized due to the strong antiferromagnetic couplings for dimer bonds. A number of extraordinary magnetic properties were observed such as magnetization plateaus, dispersionless excited states, which have stimulated further experimental⁴⁻⁹⁾ and theoretical¹⁰⁻²⁵⁾ studies. On the other hand, in the compound $\text{Nd}_2\text{BaZnO}_5$ ³⁾ the neodymium ion has a large magnetic moment $J = 9/2$, leading to an antiferromagnetically ordered state below $T_N = 2.4\text{K}$.

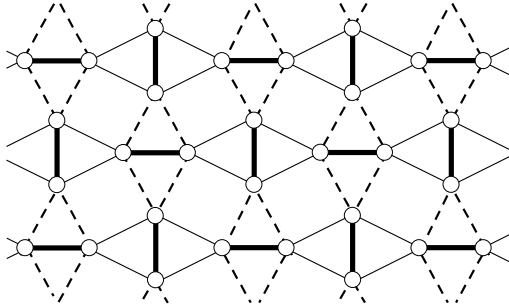


Fig. 1. Orthogonal-dimer structure: the bold, thin and broken lines represent the exchange couplings J , J' and J'' . Note that the dimers indicated by the bold lines are orthogonal to each other. When $J'' = 0$, the system is reduced to the orthogonal-dimer chain.

Magnetic properties of the above compounds may be described by the Heisenberg model on the square lattice with some diagonal bonds, which is referred to as the Shastry-Sutherland model.¹⁰⁾ This class of quantum spin models have the striking property that the direct product of independent dimers gives an exact eigenstate of the system, which can be the ground state for a certain range of parameters. Various aspects have been discussed for the $S = 1/2$ spin system, such as quantum phase transitions,¹¹⁻²¹⁾ the correlated hopping of triplet excitations,^{13,22-24)} the plateau formation in the magne-

tization curve,^{11,15,16,22,23,25)} and so on. However, magnetic properties of higher spin models ($S > 1/2$) have not been addressed so far, apart from a simple-minded approach by Shastry and Sutherland in their pioneering work.¹⁰⁾ Therefore, it is desirable to discuss how the competing exchange interactions affect the ground state properties of a higher-spin ($S > 1/2$) orthogonal-dimer model.

In a previous paper,²¹⁾ we have investigated the one-dimensional version of the orthogonal-dimer spin model with an arbitrary spin, and have shown that first-order quantum phase transitions occur ($2S$) times when the ratio of two competing antiferromagnetic exchange interactions are changed. In particular, in the $S = 1$ system, a non-trivial spin gap phase exists between the dimer and the plaquette phases. In the present paper, we investigate the ground-state phase diagram of the two-dimensional (2D) $S = 1$ Shastry-Sutherland model, and clarify how the above spin-gap phases in 1D are adiabatically connected to those in the 2D model by introducing interchain couplings.

The paper is organized as follows. In Sec. 2, we introduce the $S = 1$ Shastry-Sutherland model, and discuss how the quantum phase transitions are affected by spatially anisotropic exchange couplings. In Sec. 3, we also discuss the effect of single-ion anisotropy, which is important for some materials. A brief summary is given in Sec. 4.

2. Phase diagram of the isotropic model

We consider the generalized Shastry-Sutherland model,

$$H = \sum_{(i,j)} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + D \sum_i (S_i^z)^2, \quad (1)$$

where \mathbf{S}_i denotes the $S = 1$ spin operator on the i th site, and $J_{ij} = J, J'$ and J'' represent the intra-dimer, the inter-dimer and the inter-chain couplings, which are all assumed to be antiferromagnetic. The single-ion anisotropy is denoted as $D(< 0)$. A schematic view of the model is given in Fig. 1.

In order to see what kind of spin-gap phases or the magnetically ordered phases are realized in the 2D

Shastry-Sutherland model ($J' = J''$), we perform the exact diagonalization ($N = 4 \times 4$ system) of the above generalized spin model. We focus on the isotropic model with $D = 0$ in this section.

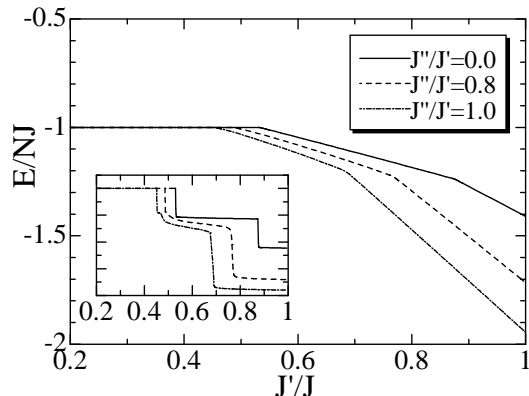


Fig. 2. Ground state energy of the isotropic model as a function of J'/J obtained by the exact diagonalization of the small system with 4×4 sites. From up to down, $J''/J' = 0.0, 0.8$ and 1.0 . Inset shows the derivative of the ground state energy, from which we can clearly see the nature of the first-order phase transition.

The ground state energy computed for the isotropic model ($D = 0$) is shown in Fig. 2. It is seen that two unambiguous cusps appear in the energy diagram, irrespective of the choice of the exchange couplings, implying that the first-order quantum phase transition occurs twice when J'/J increases. The reason why we obtain such clear cusp structures even for the small 4×4 system is closely related to the characteristic orthogonal-dimer structure, for which the direct product of spatially decoupled dimers gives an exact eigenstate, making the spin correlation length extremely short. The phase diagram determined from the cusp structure of the ground-state energy is shown in Fig. 3.

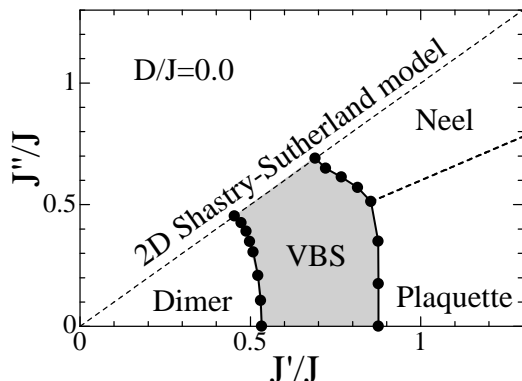


Fig. 3. Phase diagram for the isotropic model, $D = 0$. The bold lines with solid circles represent the phase boundary at which the first-order phase transition occurs. The broken line between the plaquette phase and the antiferromagnetic (AF) phase indicates the approximate location of the phase boundary.

Let us start with the case of $J'' = 0$, which is equivalent to the orthogonal-dimer spin chain studied previously.²¹⁾ In the chain model, the dimer- (plaquette-) singlet phase is realized for $J'/J < 0.56$ ($J'/J > 0.88$), both of which are characterized by the disordered ground

state with a triplet excitation gap. In the intermediate region ($0.55 < J'/J < 0.88$), another singlet phase with a triplet excitation gap is stabilized by strong frustration, which is known to be topologically equivalent to the Haldane-gap phase.²¹⁾ As is discussed in detail below, this frustration-induced state is composed of periodic alignment of dimer- and plaquette-singlets of $S = 1/2$ decoupled spins, so that it is regarded as a kind of Valence Bond Solid (VBS) state (see Fig. 4).

We naively expect that these spin-gap phases may be unstable in the presence of the interchain coupling since such quasi-one dimensional $S = 1$ spin system is usually driven to the antiferromagnetically ordered phase.^{26–31)} However, according to the present exact diagonalization study, we find that two of the spin gap phases, i.e. the dimer phase and the intermediate VBS phase, are stable against the interchain couplings, and persist even in the Shastry-Sutherland model ($J'' = J'$). In particular, it is remarkable that the nontrivial VBS phase exists even in the 2D Shastry-Sutherland model.

The above three phases are clearly distinguished from each other according to the topological nature specified by the VBS description,³²⁾ where each singlet ground state is represented by the assembly of the singlet bonds between the decomposed $S = 1/2$ spins. Shown in Fig. 4 is the VBS description of these spin gap phases. In the

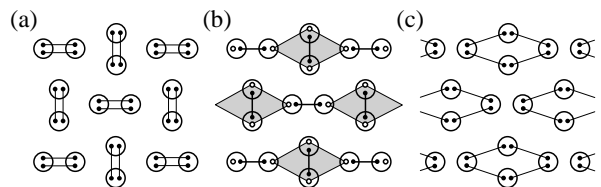


Fig. 4. VBS picture of the dimer phase (a), the frustration-induced phase (b) and the plaquette phase (c) for the generalized $S = 1$ Shastry-Sutherland model. In this description, it is assumed that the original $S = 1$ spin is decomposed into two $S = 1/2$ spins denoted by dots, and each bond connects the decoupled $S = 1/2$ spins to make singlet. Note that the system is covered by the periodic array of dimers and plaquettes for the intermediate VBS phase (b).

dimer phase, two singlet bonds are located on the strong exchange coupling J . Then the assembly of the singlet dimers gives the exact ground state of the Hamiltonian eq. (1). On the other hand, in the frustration-induced VBS phase, one of the decomposed spins at each site is connected to the nearest neighbor spin to form the singlet-dimer. Another decomposed spin is connected to other three spins to form the plaquette singlet [Fig. 4(b)]. This phase may be referred to as the "dimer-plaquette VBS phase", since it is composed of periodically alternating dimer and plaquette singlets.

Somewhat subtle is the stability of the plaquette phase against the antiferromagnetic ordered phase. Since the plaquette state forms a four-spin singlet network [Fig. 4(c)], whose wavefunction is rather extended spatially, it will show a second-order phase transition to the ordered phase as the interchain coupling is increased. This is indeed the case for the $S = 1/2$ model, for which the plaquette phase becomes unstable at a cer-

tain interchain coupling, and undergoes a second-order phase transition to the magnetically ordered phase except for a very narrow window of the choice of the exchange couplings.^{12, 19, 20} In the present $S = 1$ case, the plaquette phase is even more unstable compared with the $S = 1/2$ case since the $S = 1$ system favors the ordered state. Therefore we believe that the plaquette phase may completely disappear in the Shastry-Sutherland model ($J'' = J'$), although it is difficult to determine the phase boundary of the second-order transition numerically from the small system. The phase boundary between the plaquette phase and the antiferromagnetic phase is shown as a guide to eyes in Fig. 3.

3. Effects of single-ion anisotropy

We now discuss the effect of single-ion anisotropy, which sometimes plays an important role in stabilizing the magnetically ordered state in real materials with higher spins. It is naively expected that such anisotropy has a tendency to drive the system to the magnetically ordered phase discussed in the previous section. However, it should be noticed that the magnetic phase stabilized by single-ion anisotropy may be distinct from the magnetic phase in the previous section due to the competing interactions.

We have verified that there are indeed two kind of ordered phases in our model, which are labeled by Neel (I) and Neel (II), whose spin configuration is schematically drawn in Fig. 5. The phase (I) is the Neel ordered phase introduced above in this paper, characterized by order moments staggered among the dimers. In contrast the phase (II) corresponds to a staggering along the chains and is expected for the Neel phase stabilized by single-ion anisotropy. Obviously, the gradual increase of the interchain coupling J''/J yields an incompatible situation for the staggered ordering along the chain and the phase (I) becomes favorable.

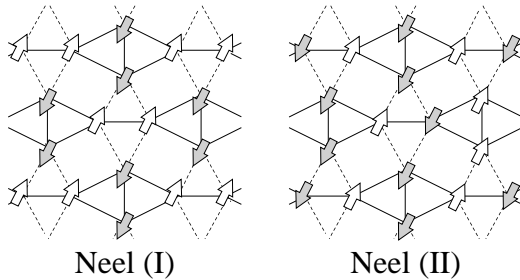


Fig. 5. Spin configurations for the Neel ordered phases (I) and (II).

The phase transition between these two distinct ordered phases should be of first-order, since there is no symmetry-hierarchy between them. To clarify this point, we make use of the Ising expansion,³³ starting from two different spin configurations shown in Fig. 5. The accuracy of this approach will be further confirmed below by comparing the results with those of the exact diagonalization calculation. To perform the series expansion, we

now divide the original Hamiltonian into two parts as

$$H = H_0 + \lambda H_1, \quad (2)$$

$$H_0 = \sum J_{ij} S_i^z S_j^z + D \sum (S_i^z)^2,$$

$$H_1 = \sum J_{ij} (S_i^x S_j^x + S_i^y S_j^y). \quad (3)$$

Then the ground state energy of each ordered state is expanded in λ as,

$$\begin{aligned} \frac{E_I}{N} &= \frac{J}{2} - J' - J'' + D - \left(\frac{J'^2}{E_{I,1}} + \frac{J''^2}{E_{I,2}} \right) \lambda^2 \\ &+ J \left[\left(\frac{J'}{E_{I,1}} \right)^2 + \left(\frac{J''}{E_{II,2}} \right)^2 \right] \lambda^3 + O(\lambda^4) \end{aligned} \quad (4)$$

$$\begin{aligned} \frac{E_{II}}{N} &= -J' + D - \left(\frac{J'^2}{E_{II,1}} + \frac{J''^2}{2E_{II,2}} + \frac{J^2}{4E_{II,3}} \right) \lambda^2 \\ &+ J \left[\left(\frac{J'}{E_{II,1}} \right)^2 + \frac{J''^2}{E_{II,2}E_{II,3}} \right] \lambda^3 + O(\lambda^4) \end{aligned} \quad (5)$$

where N is a total number of sites, $E_{I,1} = -2J + 3J' + 4J'' - 2D$, $E_{I,2} = -2J + 4J' + 3J'' - 2D$, $E_{II,1} = 3J' - 2D$, $E_{II,2} = 4J' - J'' - 2D$ and $E_{II,3} = J + 4J' - 2D$. It is found that the series coefficients in the expansion approach zero in both cases when $D \rightarrow -\infty$, from which we can see that the Ising expansion is an appropriate method to discuss the magnetic properties of the model quantitatively. We show the energy computed for each phase ($\lambda \rightarrow 1$) in Fig. 6. It is seen that two curves

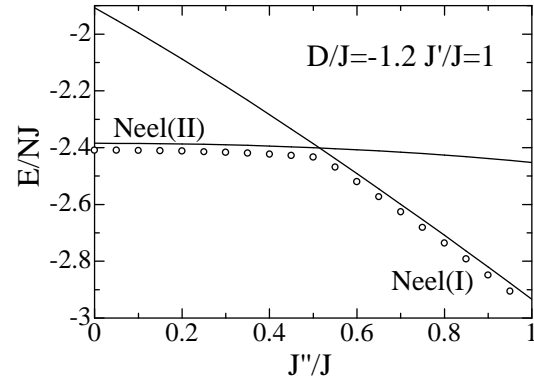


Fig. 6. Ground state energy as a function of J''/J for the anisotropic model. Solid lines are obtained by the Ising expansion up to the third order in λ . The open circles are obtained by the exact diagonalization calculation for the system of $N = 4 \times 4$.

drawn for the ground-state energy intersect each other near $J''/J \sim 0.5$, which implies that a first-order quantum phase transition occurs between the ordered phases (I) and (II). These results agree well with those obtained by the exact diagonalization, shown by open circles in Fig. 6 although there are slight differences between them due to intrinsic deviations for each method. The critical value for the first-order transition is estimated rather accurately, allowing us to determine the phase boundary.

Shown in Fig. 7 is the phase diagram thus obtained by the exact diagonalization and the Ising expansion. It

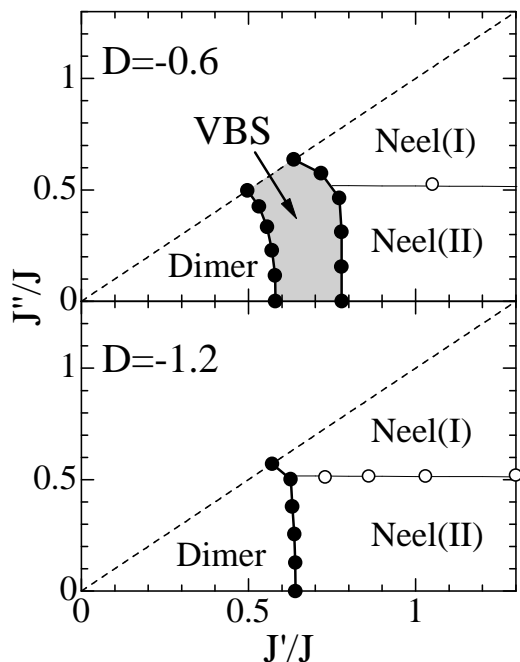


Fig. 7. The phase diagram for $D/J = -0.6$ and -1.2 . The bold lines with solid circles represent the phase boundary of the first-order transitions determined by the exact diagonalization for the 4×4 lattice. The thin solid line between the two Neel phases, which is also first-order, is determined by the third-order series expansion of the ground-state energy. The open circles are obtained by the exact diagonalization calculation.

is shown that the dimer phase and the dimer-plaquette VBS phase are stable against not only the interchain coupling but also reasonably large single-ion anisotropy ($D/J = -0.6$). However, when the anisotropy becomes larger ($D/J = -1.2$), the VBS phase eventually disappears in the phase diagram, while the dimer phase still exists. This is consistent with the fact that any VBS-like state may not be realized when the system approaches the classical limit.

As for the magnetic states, there are two distinct phases (I) and (II) separated by the first-order phase transition, as mentioned above. It is to be noted that the corresponding phase boundary is given by an almost straight line. This reflects the fact that the chosen anisotropy parameters $D/J = -0.6$ and -1.2 are considered to be rather large in the sense that the system possesses the nature expected for a classical system; the phase boundary indeed becomes exactly straight for the Ising model. The phase diagram shown above for rather large values of D possesses three or four distinct phases. We should recall here that the plaquette phase realized for $D = 0$ is completely suppressed by the presence of the magnetic phase (II) in both of the above cases. When the anisotropy parameter D decreases from these values, the plaquette phase can overcome the magnetic phase (II) and be the ground state in the region of small J''/J . The phase transition between the plaquette phase and the magnetic phase (II) may be of second order. Although we cannot determine such a second-order phase boundary correctly by means of the small-size calculation, five distinct phases should be certainly realized for

the case of small anisotropy.

The calculation presented here has been restricted to a rather small system size or lower-order perturbation. However, we believe that the above phase diagram correctly describes the ground state properties of the generalized version of the $S = 1$ orthogonal-dimer model.

4. Summary

We have investigated quantum phase transitions of the $S = 1$ Shastry-Sutherland model with single-ion anisotropy. By analyzing a generalized 2D model, we have shown how the distinct spin gap phases stabilized in the spin chain persist or disappear in the 2D Shastry-Sutherland model. The obtained phase diagram has a quite rich structure especially in the case with single ion anisotropy. We wish to particularly emphasize that the frustration-induced VBS phase exists even in the 2D Shastry-Sutherland model. If this type of the spin-gap phase can be found in real compounds, it serves as a novel example of the VBS state in 2D quantum spin systems. Similar but slightly different VBS singlet phases may be also possible, e.g. in the frustrated $S = 1$ spin systems with the Kagome lattice,^{34,35)} the pyrochlore lattice,³⁶⁻³⁸⁾ etc.

It is interesting to ask whether a higher-spin Shastry Sutherland model with $S > 1$ can still realize such a VBS state. This is an open problem to be addressed in the future study.

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